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Report:

Aerodynamic levitation combined with laser heating is a powerful and versatile technique for studying the structure, dynamics and macroscopic properties of high-temperature oxide liquids. In this experiment, we applied these techniques to study structural changes in oxide liquids at sub-second time scales as the onset of solidification – either crystallization or a glass transition - is approached.

The experiment described in this report were carried out at the CRG BM02 beam line. We have used the high temperature chamber developed at CRMHT for the BM2 beam line and described in details in ref [1]. This chamber uses aerodynamic levitation and CO_2 laser heating. A spherical sample with a diameter around 3mm is placed in a conical nozzle in the centre of the chamber. An argon gas flow is used to maintain the sample in a stable position without any contact with the nozzle. The laser beam, focused on the sample using spherical mirrors, makes it possible to reach the molten state after a few seconds. When the sample is completely melted, it is possible to turn the laser power off in order to access the free cooling of the sample. The complete solidification occurs after 1 or 2 seconds. By using sequence of measurements with an exposure time less than 0.1s, it is possible to look at the dynamical aspect of the crystallization or glass transition.

Measurements have been performed at 16.95 keV using a 120° INEL curved detector. This detector enable the measurement of the diffracted beam over a large 2Θ range simultaneously

with very short acquisition time. For this experiments, we used a counting time of 50ms for one acquisition. In order to study the glass transition, we used calcium aluminate compounds $(CaO)_x(Al_2O_3)_{1-x}$. With these materials one can obtain the glasses over a broad range of compositions and they are extremely suitable for our levitation technique.

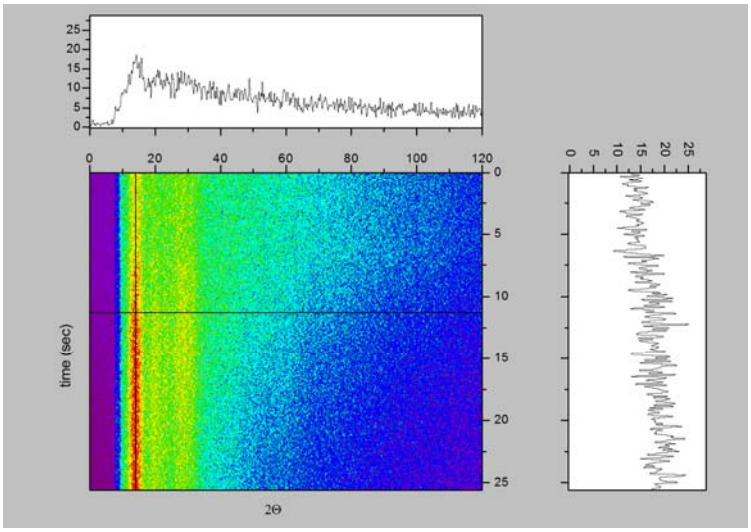

Figure 1

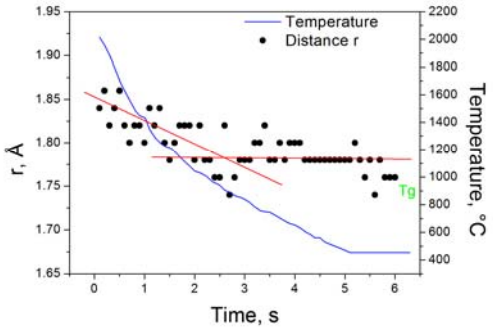

Figure 2

(right) Changes in first neighbour shell during solidification. The second scale presents the temperature evolution as a function of acquisition time

(left) The evolution in time with the temperature of $(CaO)(Al_2O_3)$ sample during free cooling

The map shown on figure 1 represents the mesureament on $(CaO)(Al_2O_3)$. It is the evolution in time of the diffraction patterns from the levitated sample that undergoes a glass transition around 910°C. After shuting down the lasers the system was cooled down freely. During the process we have done 512 acquisitions 50 ms each. Two black lines present the places where the profiles were taken. In the horisontal profile we see a typical single acquisition and in the vertical direction we can follow the evolution in time in the region of interest (first pic in our case) during each mesureaments.

The diagram presented on figure 2 shows the changes in the nearest neighbour distance in the pair distribution function calculated from measured structure factors. The second scale presents the temperature evolution as a function of acquisition time. We clearly see two regions, the shortening during the cooling of the liquid in the first part and a constant first neighbour distance in the remaining part that correponds to glass state. Red lines present linear fit of the experimental points in these two cases. The intersection of two gives the temperature of the glass transition T_g of the sample.

The data treatment is still in progress. With these measurements, it will be possible to follow more closely the evolution with temperature during free solidification of the samples. We will be able to characterize more precisely the dynamics of crystallization and/or glass transition effects during free cooling.

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1 L. Hennet, D. Thiaudière, M. Gailhanou, C. Landron, J. P. Coutures, D. L. Price, Rev. Sci. Instrum., 73, 124-129 (2002)